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THICK $YBa_2Cu_3O_{7-x}BaSnO_3$ FILMS WITH ENHANCED CRITICAL CURRENT DENSITY AT HIGH MAGNETIC FIELDS (POSTPRINT)

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14. ABSTRACT

The thickness dependence was studied for the critical current density (J_c) of $YBa_2Cu_3O_{7-x}$ (YBCO)+ $BaSnO_3$ (BSO) nanocomposite films. These films showed a significantly reduced decline of the J_c with thickness, especially at high magnetic fields. For example, a 2-μm-thick YBCO+BSO film had a J_c ~3×10^5 A/cm^2 at 5 T as compared to a typical J_c of 2.4×10^3 A/cm^2 at 5 T for a 300-nm-thick YBCO film. The thick YBCO+BSO films maintained high T_c (>88 K) and had a high density (2.5×10^{11}/cm^2) of continuous BSO nanocolumns that likely contributed for the observed J_c enhancements.

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Thick $YBa_2Cu_3O_{7-x}+BaSnO_3$ films with enhanced critical current density at high magnetic fields

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The thickness dependence was studied for the critical current density (J_c) of $YBa_2Cu_3O_{7-x}$(YBCO)+$BaSnO_3$ (BSO) nanocomposite films. These films showed a significantly reduced decline of the J_c with thickness, especially at high magnetic fields. For example, a 2 μm thick YBCO+BSO film had a $J_c \sim 3\times10^5$ A/cm^2 at 5 T as compared to a typical J_c of 2.4 $\times10^3$ A/cm^2 at 5 T for a 300 nm thick YBCO film. The thick YBCO+BSO films maintained high T_c (>88 K) and had a high density (2.5×10^{11}/cm^2) of continuous BSO nanocolumns that likely contributed for the observed J_c enhancements. © *2008 American Institute of Physics*.
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Coated conductor technology employs $YBa_2Cu_3O_{7-x}$ (YBCO) films deposited on flexible metallic substrates to manufacture high temperature superconducting electrical wires for applications such as transmission cables, power machinery, etc.[1–4] Although impressive advancements have been made in recent years to process long lengths, further enhancements in the critical current density (J_c) in high magnetic fields is still necessary. In addition, thick YBCO films (>2 μm) with a high J_c in applied magnetic fields are required to achieve a high engineering current density (J_e), especially for generators. High J_e thick films not only reduce the weight of the windings but also the associated cost of the required wire for an application. For example, a J_e of 30–40 kA/cm^2 in operating fields of 3–5 T is desired for wires to be used in generator and motor applications.[2] High field magnets desire an extension of the irreversibility field to higher values, making significant J_c increases necessary at 6–10 T or more. However, when regular or some other flux pinned YBCO films are grown, their associated J_c decreases asymptotically with increases in thickness at a given field and shows a significant decline with increases in magnetic field for a given film thickness.[5,6] Even previously announced nanoparticle pinned YBCO films show the rapid deterioration in J_c as the magnetic field is increased due to the lack of effective high-field flux pinning centers.[7,8] Several nanoparticulate pinning centers of nonsuperconducting materials such as Y_2BaCuO_5,[7] $BaZrO_3$,[8,9] $BaSnO_3$ (BSO),[10] etc., are added in the YBCO film matrix to improve the J_c and more efforts are still ongoing worldwide.[11–13]

It has been noted that the self-field J_c decreases from 4–6 to 1 MA/cm^2 as the thickness is increased to higher than 2 μm in regular YBCO films.[5] In applied magnetic fields, the J_c of 1 μm thick YBCO films decrease further, showing a J_c of $\sim10^4$ A/cm^2 at 7 T.[6] It is proposed that degradation in the degree of crystallinity, or through-thickness cation disorder level variations could be partially responsible.[14,15] One of the methods to overcome the J_c dependence on the film thickness is to grow alternate layers of CeO_2 and YBCO.[16] Although self-field J_c data were improved, a drop in J_c with the magnetic field still appears to continue. In addition, as opposed to multiple pulsed laser deposition (PLD) targets used in this approach, it is advantageous if one target is used to grow thick YBCO films. It is of interest to maintain the improvements in J_c at even higher fields, e.g., >5 T in order to make superconducting magnets for magnetic resonance imaging and superconducting magnet energy storage systems.

Another approach is to process thick YBCO+$BaZrO_3$ (BZO) films using a single premixed target.[17] Although thick films with improved J_c are reported, it was found that as the BZO content is increased, the critical transition temperature (T_c) decreases in this system, thereby limiting the amount of BZO that can be incorporated into the YBCO films.[18] Hence there is a need for a means to process thicker YBCO films (>2 μm) with higher density of nanocolumns to maintain a high J_c in high magnetic fields without a reduction in T_c. Recently 300 nm thick YBCO+BSO nanocomposite films were processed using a dual-phase sectored PLD target method without an appreciable decrease in T_c (Ref. 10) even with large amounts of BSO. YBCO+BSO thin films prepared by this method were found to have high T_c, high J_c in both $H//c$, and $H//ab$ orientations in high magnetic fields.[19] More than an order of magnitude improvement in J_c was observed in 300 nm thick film samples as compared to YBCO samples of similar thickness at 5 T, $H//c$.[10] Recently Mele *et al.*[12] compared the performance of YBCO+BZO and YBCO+BSO thin films as well and showed that the films with BSO have higher pinning force. However, thick film (>2 μm) performance in YBCO+BSO films at high fields was not previously discussed. Here we demonstrate that thick YBCO+BSO nanocomposite films with higher J_c at high magnetic fields (>5 T) than the thin YBCO+BSO and regular YBCO films can be grown if suitable microstructure is maintained.

All the films discussed in the present letter were prepared by using PLD. A dual-phase sectored PLD target approach, which is discussed in detail elsewhere,[19–21] was used to process the thick films. Essentially, a thin disk of a BSO sector is adhered to the top surface of a YBCO target and as the target is rotated during the pulsed laser deposition, BSO is introduced into a growing YBCO film. Films were grown at 780 °C using a 248 nm wavelength laser with an energy

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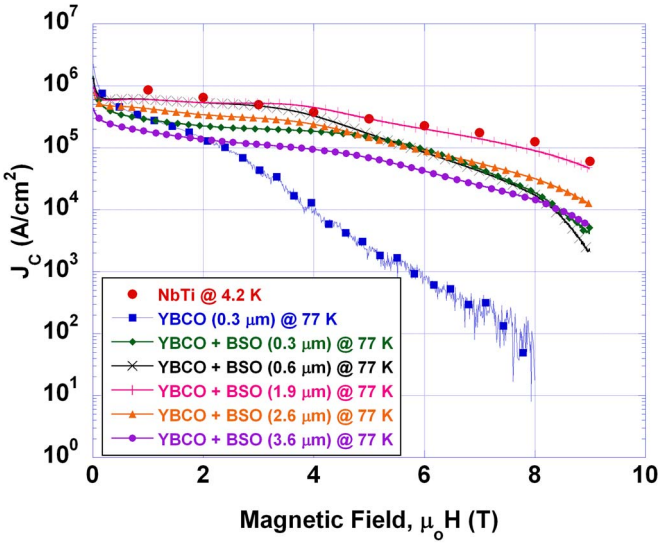

FIG. 1. (Color online) Critical current density (J_c) of different thickness YBCO+BSO nanocomposite films as compared to 300 nm YBCO samples at 77 K, $H//c$ and NbTi samples at 4.2 K.

density of 2–3 J/cm^2. Films with thicknesses varying from 300 nm to 3.6 μm were prepared on single crystal $LaAlO_3$ (100) substrates just by extending the length of deposition time from 20 min to 4 h, without any other process optimization for different thickness samples. The thickness of all the samples was measured using a KLA Tencor profilometer. The J_c was measured using the hysteresis loops data acquired from a Quantum Design PPMS vibrating sample magnetometer at 77 K in $H//c$ orientation and applying Bean's model ($J_c = 20 \times \Delta M/[a(1-a/3b)]$, where ΔM is the hysteresis in emu cm^{-3} and a and b are the sample dimensions). The samples were of square shape with $\sim(3 \times 3$ mm^2) size. Film plan view microstructures were imaged using a high resolution scanning electron microscope and cross-sectional images were obtained by using a transmission electron microscope (TEM). X-ray diffraction was done on the samples to estimate the axis-growth content in the samples.

The J_c at 77 K as a function of applied magnetic field of YBCO+BSO films with different thicknesses are compared to 300 nm thick YBCO film at 77 K and a NbTi sample at 4.2 K in Fig. 1. These films were found to contain $\sim$15–20 mol % of BSO by image analyses. It can be seen that the J_c at 9 T in 1.9 μm thick YBCO+BSO films is nearly three orders of magnitude higher as compared to 300 nm thick YBCO film and is almost similar to NbTi. No significant degradation was noticed in YBCO+BSO films when the thickness is increased from 0.3 to 3.6 μm. In fact, as compared to YBCO+BSO films of 300 nm thickness, the 2.6 μm thick films had J_c that is more than five times higher over a range of fields. Contrary to what is observed in regular YBCO, as the thickness is increased, the films had some initial improvement. In addition, the 2 μm thick YBCO +BSO film ($J_c > 0.1$ MA/cm^2) showed more than an order of magnitude increase in J_c at 7 T as compared to the J_c of a 1 μm thick regular YBCO film ($J_c = 0.01-0.02$ MA/cm^2) reported in literature.[6] The J_c at 8 T of the 3.6 μm thick film is almost the same as the 300 nm thick YBCO+BSO nanocomposite film showing nearly no loss of J_c with thickness when measured at this field. Although the J_c of 3.6 μm thick films is slightly lower than that of the 2 or 2.6 μm thick films, an order of magnitude improvement in the J_c at 4 T in a 3.6 μm thick film as compared to regular YBCO film can also be observed. With the present processing conditions, films with thickness around 2–2.5 μm were found to have better quality than the films with other thickness. An increased extent of a-axis grain growth in the films was determined from both x-ray diffraction and scanning electron micrograph (SEM) observations as the thickness was increased especially in films with thickness greater than 2 μm. This increased a-axis grain growth is thought to be partially responsible for the reduction in J_c in 3.6 μm thick film. It may be contended that the in-field J_c of the films did not appreciably decrease with thickness from 300 nm to 2.5 μm since the self-field J_c of the films was lower to begin with. However, this is not the case since the in-field starting values are cited above as being significantly improved above that of other YBCO films. It is noted that the self-field values are slightly depressed, but for in-field applications this is irrelevant since the values are much improved in field and maintain the high values up to large fields, which is what matters for the applications. The self-field J_c is lower due to high volume fraction BSO content in the YBCO+BSO nanocomposite films, where pinning centers play a greatly reduced role.

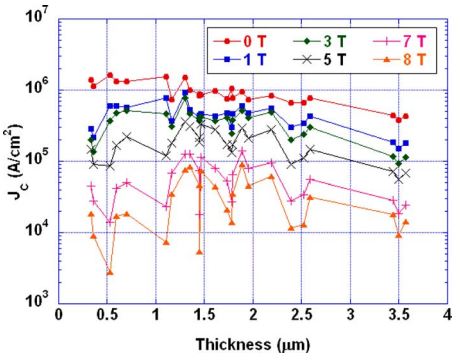

FIG. 2. (Color online) J_c values of several YBCO+BSO films with different thicknesses at different applied fields measured at 77 K, $H//c$ orientation.

Of particular importance is that the J_c reported in this letter are derived from the magnetization measurements that are traditionally known to give lower J_c data than the transport current measurements. However, by comparing the J_c of 300 nm thick films with the 3.6 μm film measured in the same instrument, it is clear that the J_c remains high at high fields and does not show significant degradation as the thickness is increased at magnetic fields up to 9 T, the limit of our particular magnetometer model. The incorporation of a high volume fraction of ($\sim$10 vol %) of nonsuperconducting BSO phase would with no pinning enhancement reduce the J_c due to reduction of the superconducting phase (by $\sim$10 vol %), indicating the additional strength of the pinning centers. A T_c of 87–88.5 K was measured in these thick YBCO+BSO films. Although high volume content of BSO was added, it did not reduce T_c as opposed to BZO, which reduces the T_c with >2 vol %.[18]

To see the J_c thickness dependence clearly, the J_c at 77 K in several of the YBCO+BSO nanocomposite films is plotted as a function of their thickness in Fig. 2. It can be seen that up to $\sim$2.5 μm, J_c values at fields greater than 1 T are higher in thick films than the corresponding thin films (300 nm) at any given field except self-field. It should be noted

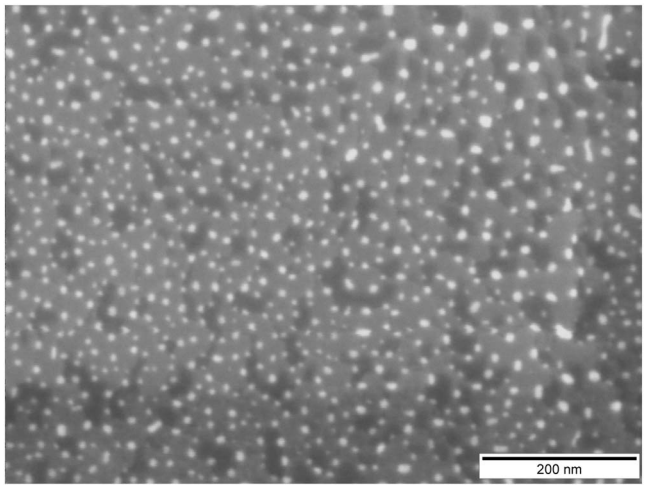

FIG. 3. SEM photomicrograph of a ~3.6 μm film showing uniform distribution of BSO in YBCO matrix.

again that self-field J_c values are lower due to the high volume fraction of the BSO as discussed above. Further, the magnitude of drop in J_c with a change in field from 0 to 8 T is lesser in YBCO+BSO thicker films as evidenced by the narrower J_c data spread in 3.6 μm thick sample compared to 300 nm thick YBCO+BSO sample in Fig. 2.

Figure 3 shows a plan view SEM of a 3.6 μm thick YBCO+BSO film showing uniform distribution of BSO (the bright phase) in these films. Figure 4 shows a cross-sectional TEM image of another sample of similar thickness. BSO nanocolumns were found to be straight, continuous, and extend all the way through the thickness of the films. The average column size was found to be 10–11 nm and the separation was nearly 20 nm. The area density is calculated to be $2.5\times10^{11}/\mathrm{cm}^2$ from these micrographs. Other defects such as dislocations, strain fields, and stacking faults were also found to be present in thick films similar to the nanocolumns and defects observed in previously reported 300 nm thick YBCO+BSO films.[19] The mechanism for such a microstructure formation is believed to be due to the strain energy minimization that exists due to the lattice mismatch (7.7%) between BSO and YBCO. A high density of continuous, solid BSO nanocolumns and associated defects, and increased irreversibility field are responsible for the improved properties observed in YBCO+BSO films. What is shown in this letter is that a microstructure with long BSO nanocolumns can be maintained in thick films and that these nanocolumns help to grow thicker films that can maintain high J_c at high magnetic fields.

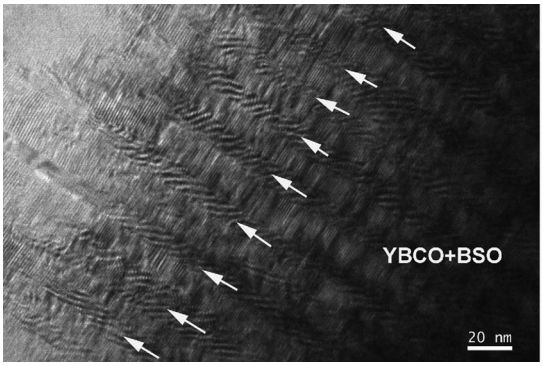

FIG. 4. TEM cross-sectional image of a ~3.6 μm film showing the BSO nanocolumns extending throughout the thickness of the films.

In conclusion, it is shown that YBCO+BSO films formed by a dual-sector pulsed laser ablation method demonstrate maintenance of high J_c in thick films at high magnetic fields. More than two orders of magnitude improvement at 9 T is possible with a high volume percent of BSO in the thicker films in the form of nanocolumns as compared to 300 nm thick regular YBCO. However, process optimization in terms of increased deposition temperature or laser energy, deposition rate, etc., may further improve the properties of these thick films, especially beyond 2.6 μm.

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